

Raman transitions: Adiabatic elimination revisited

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Abstract. In multi-level systems, the commonly used adiabatic elimination is a method for approximating the dynamics of the system by eliminating irrelevant, non-resonantly coupled levels. This procedure is, however, somewhat ambiguous and it is not clear how to improve on it systematically. We use an integro-differential equation for the probability amplitudes of the levels of interest, which is equivalent to the original Schrödinger equation for all probability amplitudes. In conjunction with a Markov approximation, the integro-differential equation is then used to generate a hierarchy of approximations, in which the zeroth order is the adiabatic-elimination approximation. It works well with a proper choice of interaction picture; the procedure suggests criteria for optimizing this choice. The first-order approximation in the hierarchy is found to be sufficient for practical purposes, and is not so sensitive to the choice of interaction picture. We illustrate the method for a single three-level atom and a pair of three-level atoms with Rydberg blockade.

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1. Introduction

The coherent manipulation of quantum states is central to a large variety of contemporary research in physics, among them experiments that aim at processing quantum information. When implementing a qubit in the internal degrees of freedom of a trapped atom or ion, the relevant quantum states are different electronic energy levels manipulated by a combination of static and time-dependent electromagnetic fields. The ideal case of a two-level system resonantly driven by a laser is rarely available as the levels of interest may not be coupled directly by a dipole transition, or the transition occurs at an inconvenient wavelength. Instead, two levels are often coupled indirectly through

intermediate levels, leading to a multi-level system, which can be very complicated to analyze exactly. Fortunately, the intermediate levels are usually barely populated and are of little consequence, other than providing a coupling route, to the dynamics of the levels of interest. It is then common to employ the procedure of adiabatic elimination to remove these irrelevant levels, thereby achieving a considerable simplification of the formalism in terms of a lower-dimensional effective description.

In this paper we first review the standard method of adiabatic elimination and the problems that may arise when applying it (Section 2). Then, in Section 3, we present a systematic approach for separating the more relevant and the less relevant subspaces by means of an integro-differential equation of Lippmann-Schwinger type that can be used to generate a hierarchy of approximations, starting with the usual adiabatic elimination. A technical point of considerable importance is the choice of an appropriate interaction picture; we propose optimality criteria for a judicious choice. In Section 4 finally, we use a driven Raman transition of Λ -type in a three-level atom, and also two atoms with a three-level cascade experiencing a Rydberg blockade, for case studies that illustrate the usefulness of our approach.

2. Brief review of adiabatic elimination

2.1. Example: Raman transition in a three-level system

In a three-level system, stimulated Raman transition is a two-photon process used to couple a *ground state* $|0\rangle$ to a *target state* $|1\rangle$ via a far-detuned *intermediate state* $|e\rangle$, with respective energies $\hbar\omega_0$, $\hbar\omega_1$, and $\hbar\omega_e$. The use of such an indirect coupling is necessary whenever a direct transition between the ground state and the target state is not possible, because the transition is dipole forbidden, or has a transition frequency outside the range accessible to common lasers. Depending on the energies of the three levels, the system is either in the Λ configuration ($\omega_0, \omega_1 < \omega_e$), the V configuration ($\omega_0, \omega_1 > \omega_e$), or the cascade configuration ($\omega_0 < \omega_e < \omega_1$).

To be specific, we will focus on the Λ configuration of figure 1, but the other configurations can be dealt with in very much the same way. The transitions are driven by two distinguishable lasers (with different frequencies or polarizations, for example) of frequencies ω_{L0} and ω_{L1} . The i th laser has a detuning $\Delta_i = \omega_e - \omega_i - \omega_{Li}$ to the intermediate state. It is convenient to introduce the average detuning $\Delta = \frac{1}{2}(\Delta_0 + \Delta_1)$, as well as the overall detuning $\delta = \Delta_0 - \Delta_1$ of the two-photon process.

After applying the dipole and rotating-wave approximations, the Hamiltonian of the three-level system is represented in the basis $\{|0\rangle, |1\rangle, |e\rangle\}$ by the matrix

$$H = \hbar \begin{pmatrix} 0 & 0 & \frac{1}{2}\Omega_0 e^{i\omega_{L0}t} \\ 0 & \omega_1 & \frac{1}{2}\Omega_1 e^{i\omega_{L1}t} \\ \frac{1}{2}\Omega_0^* e^{-i\omega_{L0}t} & \frac{1}{2}\Omega_1^* e^{-i\omega_{L1}t} & \omega_e \end{pmatrix}, \quad (1)$$

where the Rabi frequencies Ω_i depend on the amplitude and the polarization of the electric field, as well as the dipole matrix elements between the states. In the interaction

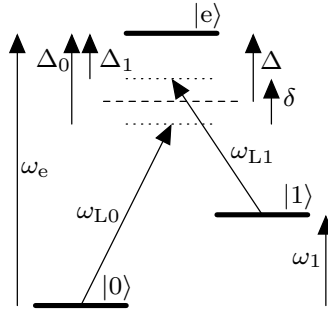


Figure 1. The average detuning Δ is large compared with the Rabi frequencies of the lasers with frequencies ω_{L0} and ω_{L1} that drive the transitions $|0\rangle \rightarrow |1\rangle$ and $|0\rangle \rightarrow |1\rangle$, respectively. The overall detuning δ of the two-photon transition is small.

picture defined by the splitting

$$H = H_0 + H_1 \quad \text{with} \quad H_0 = \hbar \begin{pmatrix} \frac{1}{2}\delta & 0 & 0 \\ 0 & \omega_{L0} + \omega_{L1} + \frac{1}{2}\delta & 0 \\ 0 & 0 & \omega_{L0} + \frac{1}{2}\delta \end{pmatrix}, \quad (2)$$

the resulting Hamiltonian is time-independent,

$$H_1 = e^{iH_0 t/\hbar} H_1 e^{-iH_0 t/\hbar} = \frac{\hbar}{2} \begin{pmatrix} -\delta & 0 & \Omega_0 \\ 0 & \delta & \Omega_1 \\ \Omega_0^* & \Omega_1^* & 2\Delta \end{pmatrix}, \quad (3)$$

where we indicate the splitting of the 3×3 matrix into the top-left 2×2 matrix for the relevant levels $|0\rangle$ and $|1\rangle$ and the rest that involves the intermediate level $|e\rangle$.

The time evolution of such a three-state problem can, of course, be studied using an exact eigen-decomposition, but the actual expressions for the eigenvalues and eigenvectors of H_1 are often involved and not transparent. Therefore, adiabatic elimination is frequently used to remove the intermediate state from the description and thus reduce the three-level system to a two-level system of the relevant ground and target states, with a 2×2 matrix for its effective Hamiltonian. Then all the familiar tools for two-level systems are applicable, and one can easily work out the frequency and amplitude of the oscillatory probability amplitudes as well as other details of interest.

Adiabatic elimination relies on the fact that the intermediate level, which is not populated initially, is only weakly and non-resonantly coupled to the ground state and the target state, i.e., $|\Omega_i| \ll |\Delta|$. It is then customary to set, in the equations of motion in the interaction picture,

$$i \frac{\partial}{\partial t} c_e(t) = \frac{1}{2} \Omega_0^* c_0(t) + \frac{1}{2} \Omega_1^* c_1(t) + \Delta c_e(t) \approx 0, \quad (4)$$

where c_0 , c_1 , and c_e denote the interaction-picture probability amplitudes for the respective three levels depicted in figure 1. Under this approximation, $c_e(t)$ can be expressed as a linear combination of $c_0(t)$ and $c_1(t)$, which then permits the elimination of $c_e(t)$

from the equations of motion for the probability amplitudes $c_0(t)$ and $c_1(t)$. This leads to an effective two-dimensional Hamiltonian

$$H_{\text{eff}} = -\frac{\hbar}{2} \begin{pmatrix} \delta + \frac{|\Omega_0|^2}{2\Delta} & \frac{\Omega_0\Omega_1^*}{2\Delta} \\ \frac{\Omega_1\Omega_0^*}{2\Delta} & -\delta + \frac{|\Omega_1|^2}{2\Delta} \end{pmatrix}, \quad (5)$$

now written in the basis $\{|0\rangle, |1\rangle\}$ involving the relevant states only.

2.2. Problems with adiabatic elimination

Despite its popular use in simplifying multi-level problems, the procedure of adiabatic elimination has certain ambiguities and potential problems. We present these in the form of four questions:

- (a) The basic assumption that the time derivative of $c_e(t)$ is small and can be treated as approximately vanishing is questionable. Even if the population in the intermediate state is small, its time derivative need not be small; in fact, it is rather sizeable because the large detuning makes $c_e(t)$ oscillate rapidly. *Is there a better justification for this assumption?*
- (b) There are problems with the normalization of the wave function. Originally, we have $|c_0(t)|^2 + |c_1(t)|^2 + |c_e(t)|^2 = 1$. As H_{eff} is hermitian, the two-component wave function that goes with it has $|c_0(t)|^2 + |c_1(t)|^2 = \text{constant}$. Combined with the initial condition $(c_0, c_1, c_e) = (1, 0, 0)$ this implies $c_e(t) = 0$ for all t , whereas the basic approximation in (4) says otherwise. Thus it fails to make a consistent statement about the population in the intermediate state. *Is it possible to estimate the population in the eliminated state and comply with the normalization conditions?*
- (c) It is always possible to go to another interaction picture by subtracting a multiple of the identity from H_0 , and split the Hamiltonian differently, $H = (H_0 - E) + (H_1 + E) = \tilde{H}_0 + \tilde{H}_1$. This yields a different interaction-picture Hamiltonian, $\tilde{H}_1 = H_1 + E$. The modification appears innocuous, but it results in a different effective Hamiltonian upon the adiabatic elimination of the intermediate level. There have been attempts to identify the “correct” choice of interaction picture, which gave valuable insights [1], but a definite answer is still lacking. *How can one choose an optimal interaction picture?*
- (d) For larger Rabi frequencies or smaller detunings, one observes that the adiabatic elimination does not give a trustworthy approximation. *Is there a systematic way of improving the accuracy of the adiabatic-elimination approximation?*

We offer answers to these four questions in the next sections.

3. General method

3.1. Integro-differential equation for the relevant states

In the more general situation of a multi-level system with $m + n$ energy levels, we distinguish the set of m *relevant states*, whose probability amplitudes are collected in the m -component column $\psi(t)$, and the set of n *auxiliary states*, for which we have the n -component column $\epsilon(t)$, with weak or highly non-resonant coupling between states from different sets. In the spirit of adiabatic elimination, we wish to have an effective description for the relevant states described by $\psi(t)$.

It is most convenient to work in an interaction picture where the Hamiltonian is time-independent. For a basis where the first m elements correspond to the relevant states and the following n elements to the auxiliary states, the interaction-picture Hamiltonian is composed of the submatrices ω , Ω , and Δ ,

$$H_I = \hbar \begin{pmatrix} \omega & \frac{1}{2}\Omega \\ \frac{1}{2}\Omega^\dagger & \Delta \end{pmatrix}, \quad \text{and} \quad \Psi(t) = \begin{pmatrix} \psi(t) \\ \epsilon(t) \end{pmatrix} \quad (6)$$

is the $(m + n)$ -component column of probability amplitudes. The Schrödinger equation then takes the form

$$\begin{aligned} i \frac{\partial}{\partial t} \psi(t) &= \omega \psi(t) + \frac{1}{2} \Omega \epsilon(t), \\ i \frac{\partial}{\partial t} \epsilon(t) &= \frac{1}{2} \Omega^\dagger \psi(t) + \Delta \epsilon(t). \end{aligned} \quad (7)$$

If we were using adiabatic elimination, we would now set $\frac{\partial \epsilon}{\partial t} = 0$. Instead, we solve the differential equation for $\epsilon(t)$ with the initial condition of no population in the auxiliary states, $\epsilon(t = 0) = 0$,

$$\epsilon(t) = -\frac{i}{2} \int_0^t dt' e^{-i\Delta(t-t')} \Omega^\dagger \psi(t'). \quad (8)$$

We then use this in the differential equation for $\psi(t)$ to arrive at an integro-differential equation of Lippmann-Schwinger type,

$$i \frac{\partial}{\partial t} \psi(t) = \omega \psi(t) - \frac{i}{4} \Omega \int_0^t dt' e^{-i\Delta(t-t')} \Omega^\dagger \psi(t'). \quad (9)$$

Together with (8), this is fully equivalent to the pair of equations in (7). While no approximation entered in the transition from (7) to (9), the integro-differential equation now serves as the starting point for the generation of a hierarchy of approximations. This approach is similar in spirit to, but differs in detail from, the Green's-function approach discussed in [1].

3.2. Markov approximation

The usual (zeroth-order) Markov approximation assumes that memory effects are negligible, so that $\psi(t') \approx \psi(t)$ in the integral in (8). This assumption is valid, provided that $\psi(t)$ oscillates much more slowly than $e^{-i\Delta t}$. This happens when $|\Omega|, |\omega| \ll |\Delta|$, which is the case when the auxiliary states are coupled weakly and non-resonantly to the relevant states. As discussed in standard texts, such as references [2] and [3], a coarse graining is then appropriate in the evaluation of the remaining integral over t' because time scales of order Δ^{-1} are not resolved and $e^{-i\Delta t}$ averages out over the coarse-grained time intervals. Accordingly, we have

$$\int_0^t dt' e^{-i\Delta(t-t')} = \frac{1 - e^{-i\Delta t}}{i\Delta} \approx \frac{1}{i\Delta}, \quad (10)$$

and thus obtain the effective Hamiltonian for the zeroth-order Markov approximation,

$$H_{\text{eff}}^{(0)} = \hbar \left(\omega - \Omega \frac{1}{4\Delta} \Omega^\dagger \right). \quad (11)$$

This happens to be exactly the effective Hamiltonian that adiabatic elimination would yield. In this sense, then, we have given an answer to question (a) in Section 2.2. Setting $\frac{\partial}{\partial t} c_e(t) \approx 0$ in the adiabatic elimination procedure amounts to a shorthand for the above coarse graining in time to discard rapidly oscillating features and retain only the slowly varying dynamics.

To gain some information about the evolution of $\epsilon(t)$, and so arrive at an answer to question (b), we consider the first-order Markov approximation that takes a bit of the history of the relevant states into account in the integral in (8). We accomplish this by means of the approximation

$$\psi(t') \approx \psi(t) - (t - t') \frac{\partial \psi(t)}{\partial t}, \quad (12)$$

from a Taylor-series expansion about $t' = t$. With (10) and

$$\int_0^t dt' (t - t') e^{-i\Delta(t-t')} = i \frac{\partial}{\partial \Delta} \int_0^t dt' e^{-i\Delta(t-t')} \approx -\frac{1}{\Delta^2}, \quad (13)$$

this amounts to

$$i \frac{\partial \psi(t)}{\partial t} = \left(\omega - \Omega \frac{1}{4\Delta} \Omega^\dagger \right) \psi(t) - i \Omega \frac{1}{4\Delta^2} \Omega^\dagger \frac{\partial \psi(t)}{\partial t}. \quad (14)$$

In defining the effective Hamiltonian, we now have a choice between

$$i \frac{\partial \psi(t)}{\partial t} = \left(1 + \Omega \frac{1}{4\Delta^2} \Omega^\dagger \right)^{-1} \left(\omega - \Omega \frac{1}{4\Delta} \Omega^\dagger \right) \psi(t) = \frac{1}{\hbar} H_{\text{eff}}^{(1)} \psi(t) \quad (15)$$

and

$$\begin{aligned} i \frac{\partial \psi'(t)}{\partial t} &= \left(1 + \Omega \frac{1}{4\Delta^2} \Omega^\dagger \right)^{-1/2} \left(\omega - \Omega \frac{1}{4\Delta} \Omega^\dagger \right) \left(1 + \Omega \frac{1}{4\Delta^2} \Omega^\dagger \right)^{-1/2} \psi'(t) \\ &= \frac{1}{\hbar} H_{\text{eff}}^{(1')} \psi'(t) \end{aligned} \quad (16)$$

with

$$\psi'(t) = \left(1 + \Omega \frac{1}{4\Delta^2} \Omega^\dagger\right)^{1/2} \psi(t). \quad (17)$$

The effective Hamiltonian in (16) describes the evolution of the scaled wave function $\psi'(t)$, and is hermitian in the usual sense, so that

$$\psi'(t)^\dagger \psi'(t) = \psi(t)^\dagger \left(1 + \Omega \frac{1}{4\Delta^2} \Omega^\dagger\right) \psi(t) \quad (18)$$

is constant in time. By contrast, the effective Hamiltonian in (15) is hermitian only for the modified inner product

$$(\psi_1, \psi_2) \equiv \psi_1^\dagger \left(1 + \Omega \frac{1}{4\Delta^2} \Omega^\dagger\right) \psi_2, \quad (19)$$

giving the right-hand side of (18) for the normalization of $\psi(t)$ in accordance with $(\psi, \psi) = 1$. It does not matter which formulation we prefer for the first-order Markov approximation, either (15) with (19) or (16) with (17) and (18) is fine. The two formulations are related to each other by the similarity transformation afforded by $(1 + \frac{1}{4}\Omega\Delta^{-2}\Omega^\dagger)^{1/2}$, and reliable results can only be expected if this operator does not differ much from the identity. This gives a more precise meaning to the requirement that the coupling part Ω of the interaction-picture Hamiltonian in (6) should be small on the scale set by the energy part Δ of the auxiliary levels.

A comparison of (18) with the normalization of the full column Ψ of (6),

$$\Psi^\dagger \Psi = \psi^\dagger \psi + \epsilon^\dagger \epsilon = 1, \quad (20)$$

reveals that $\epsilon^\dagger \epsilon$ is here approximated by

$$\epsilon^\dagger \epsilon \approx \psi^\dagger \Omega \frac{1}{4\Delta^2} \Omega^\dagger \psi. \quad (21)$$

This is just what the zeroth-order Markov approximation says,

$$\epsilon(t) = -\frac{i}{2} \int_0^t dt' e^{-i\Delta(t-t')} \Omega^\dagger \psi(t) \approx -\frac{1}{2\Delta} \Omega^\dagger \psi(t). \quad (22)$$

This observation is reassuring and provides the answer to question (b) in Section 2.2.

The first-order Markov approximation offers a correction to adiabatic elimination. By including another term in the Taylor-series approximation (12), one gets a second-order Markov approximation. A second time derivative of $\psi(t)$ will appear, but it can be approximated by replacing one of the time derivatives by an application of the effective Hamiltonian, thereby obtaining a Schrödinger-type first-order differential equation for $\psi(t)$. Successive terms in the Taylor series give a systematic hierarchy of approximations, with adiabatic elimination as the zeroth-order procedure, and this offers an answer to question (d). As we shall see shortly, however, the first-order Markov approximation is already of sufficient quality, so that the complications of higher-order approximations are hardly worth the trouble.

3.3. Choice of interaction picture

There still remains question (c) about the choice of a good, or perhaps optimal, interaction picture that leads to an effective Hamiltonian with the correct physics. Since we prefer interaction-picture Hamiltonians that are constant in time, Hamiltonians for different choices of interaction picture can only differ by a multiple of the identity. The effective Hamiltonians $\tilde{H}_{\text{eff}}^{(0)}$ and $\tilde{H}_{\text{eff}}^{(1)}$ of (11) and (15), respectively, in an alternate interaction picture, would therefore take the form

$$\begin{aligned}\tilde{H}_{\text{eff}}^{(0)} &= \hbar \left(\omega + \tilde{\omega} - \Omega \frac{1}{4(\Delta + \tilde{\omega})} \Omega^\dagger \right), \\ \tilde{H}_{\text{eff}}^{(1)} &= \left(1 + \Omega \frac{1}{4(\Delta + \tilde{\omega})^2} \Omega^\dagger \right)^{-1} \tilde{H}_{\text{eff}}^{(0)},\end{aligned}\tag{23}$$

where $\tilde{\omega} = E/\hbar$ for the energy parameter E in question (c).

We recall that the crucial assumption in the Markov approximation is that $\psi(t)$ oscillates slowly in comparison with $e^{-i\Delta t}$, which is invoked when replacing $\psi(t')$ by $\psi(t)$ on the right-hand side of (12) in the integro-differential equation (9). Consistency, therefore, requires that the magnitudes of the eigenvalues of the effective Hamiltonian should be as small as possible because they determine the frequencies contained in $\psi(t)$. The following three conditions for choosing the value of $\tilde{\omega}$ suggest themselves:

$$(a) \text{tr}\{\omega + \tilde{\omega}\} = 0, \quad (b) \|\tilde{H}_{\text{eff}}\|_{\text{op}} \text{ is minimal}, \quad (c) \|\tilde{H}_{\text{eff}}\|_{\text{tr}} = \text{tr}|\tilde{H}_{\text{eff}}| \text{ is minimal}.\tag{24}$$

Here, $\|H\|_{\text{op}}$ refers to the operator norm of H , while $\|H\|_{\text{tr}}$ is the trace norm. Conditions (24b) and (24c) insist on choices of $\tilde{\omega}$ such that \tilde{H}_{eff} has eigenvalues that are small in two different senses: The operator-norm condition requires minimizing the largest absolute value of the eigenvalues of \tilde{H}_{eff} ; the trace-norm condition requires minimizing the sum of the absolute values of the eigenvalues of \tilde{H}_{eff} . Condition (24a) prescribes distributing the eigenvalues of the ω part of the interaction-picture Hamiltonian (6) about 0. This does not necessarily guarantee small eigenvalues, but for the Hamiltonians in (23), a different choice of $\tilde{\omega}$ gives, as the dominant change, only an overall shift in the eigenvalues of the Hamiltonian. Condition (24a) is hence a simple alternative to a condition based on norms of \tilde{H}_{eff} , and occurs in the context of adiabatic elimination — the ω of the interaction-picture Hamiltonian H_I in (3) that gave the effective Hamiltonian (5) from adiabatic elimination satisfies condition (24a). The example in Section 4.1 provides some evidence in favor of conditions (24b) and (24c), with minimal difference between them, but the quality of the approximation is not substantially worse if condition (24a) is enforced.

4. Approximation of Three-Level Systems

4.1. One atom

In order to check the validity of, and illustrate the differences between, the adiabatic-elimination approximation and the first-order Markov approximation, we apply them to

a three-level system of the kind depicted in figure 1. For a quantitative statement about the quality of the approximations, we compare the resulting Rabi frequency Ω_R for population transfer between $|0\rangle$ and $|1\rangle$. Here, the exact $\hbar\Omega_R$ is the smallest spacing between the eigenvalues of H_I , and is approximated by the difference of the two eigenvalues of the \tilde{H}_{eff} under consideration. We first consider the case of zero overall detuning. Expanding the solutions for $\delta = 0$ to order x^4 , where $x = \frac{1}{4}(|\Omega_0|^2 + |\Omega_1|^2)/\Delta^2$, gives

$$\begin{aligned}
\text{exact solution:} \quad & \frac{1}{\Delta}\Omega_R = x - x^2 + 2x^3 - 5x^4 + O(x^5) = \frac{1}{2}(\sqrt{1+4x} - 1), \\
\text{0th-order, (24a):} \quad & \frac{1}{\Delta}\Omega_R = x, \\
\text{0th-order, (24b):} \quad & \frac{1}{\Delta}\Omega_R = x - \frac{1}{2}x^2 + \frac{1}{2}x^3 - \frac{5}{8}x^4 + O(x^5) = \sqrt{1+2x} - 1, \\
\text{0th-order, (24c):} \quad & \frac{1}{\Delta}\Omega_R = x - x^2 + 2x^3 - 5x^4 + O(x^5) = \frac{1}{2}(\sqrt{1+4x} - 1), \quad (25) \\
\text{1st-order, (24a):} \quad & \frac{1}{\Delta}\Omega_R = x - x^2 + x^3 - x^4 + O(x^5) = \frac{x}{1+x}, \\
\text{1st-order, (24b):} \quad & \frac{1}{\Delta}\Omega_R = x - x^2 + \frac{7}{4}x^3 - \frac{15}{4}x^4 + O(x^5), \\
\text{1st-order, (24c):} \quad & \frac{1}{\Delta}\Omega_R = x - x^2 + x^3 - x^4 + O(x^5) = \frac{x}{1+x}.
\end{aligned}$$

Regardless of the choice of conditions (24a)-(24c), the zeroth-order Markov approximation gives the correct Rabi frequency up to order x , while the first-order solutions give accuracy up to order x^2 . With a judicious choice of interaction picture, one can improve the approximations — for example, under the zeroth-order Markov approximation, employing condition (24c) in fact gives the *exact* Rabi frequency. However, we believe this is a coincidence rather than a general fact (see, for instance, the $\delta \neq 0$ case discussed below). A more reliable conclusion one can draw is that the first-order Markov approximation gives better accuracy, and that whether we choose condition (24a), (24b), or (24c) matters little. In fact, one might even consider minimizing $\|\tilde{H}_{\text{eff}}\|$ for other norms (for example, the Hilbert-Schmidt norm). The near-equivalence between the conditions is reassuring, since very often, condition (24a) is the easiest among the three possibilities to apply. It also validates the common choice of condition (24a) in standard adiabatic elimination.

For comparison, we consider a case where $\delta \neq 0$. In many experiments targeting full population transfer between the ground state and the target state, δ is adjusted to compensate for the light shifts of the atomic levels, due to the presence of the lasers, to give a resonant two-photon transition. Often, the choice is $\delta = (|\Omega_1|^2 - |\Omega_0|^2)/4\Delta$, a value designed to make the diagonal entries of the effective Hamiltonian (5) from adiabatic elimination equal. This ensures a resonant process, to the accuracy justified by the adiabatic-elimination approximation. For this value of δ , the predicted Rabi frequencies for the exact solution and for the zeroth-order Markov approximation under

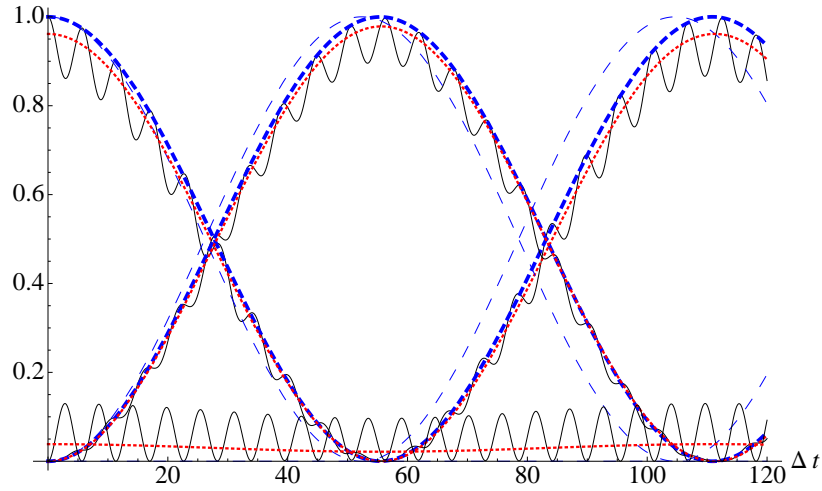


Figure 2. Population probabilities for a single-atom Raman transition in a three-level system as a function of Δt , for Rabi frequencies $\Omega_0 = 0.4\Delta$ and $\Omega_1 = 0.3\Delta$, and overall detuning $\delta = (|\Omega_1|^2 - |\Omega_0|^2)/4\Delta$. The curves that start close to 1 show the population of the ground state $|0\rangle$; those that start at 0 and increase to about 1 before decreasing again are for the target state $|1\rangle$; those that start near 0 and never grow to large values are for the intermediate state $|e\rangle$. The solid black curves plot the exact solution; the thin dashed blue curves show the zeroth-order Markov approximation with condition (24a) (standard adiabatic elimination); the thick dashed blue curves also show the zeroth-order Markov approximation, but with condition (24c); the dotted red curves are for the first-order Markov approximation with condition (24a).

the different conditions are

$$\begin{aligned}
 \text{exact solution:} \quad & \frac{1}{\Delta}\Omega_R = \sqrt{1 - \alpha^2} \{x - x^2 + [2 + O(\alpha^2)]x^3 + O(x^4)\}, \\
 \text{0th-order, (24a):} \quad & \frac{1}{\Delta}\Omega_R = \sqrt{1 - \alpha^2} x, \\
 \text{0th-order, (24b):} \quad & \frac{1}{\Delta}\Omega_R = \sqrt{1 - \alpha^2} \left\{ x - \frac{1}{2}x^2 + \left[\frac{1}{2} + O(\alpha^2) \right] x^3 + O(x^4) \right\}, \\
 \text{0th-order, (24c):} \quad & \frac{1}{\Delta}\Omega_R = \sqrt{1 - \alpha^2} \{x - [1 + O(\alpha^2)]x^2 + [2 + O(\alpha^2)]x^3 + O(x^4)\},
 \end{aligned} \tag{26}$$

where we have defined the ratio $\alpha \equiv \frac{|\Omega_0|^2 - |\Omega_1|^2}{|\Omega_0|^2 + |\Omega_1|^2} = 0$. As in the $\delta = 0$ example in (25), the zeroth-order approximation gives the correct answer to linear order in x , regardless of the choice of condition. That condition (24c) gives the exact Rabi frequency is no longer true here, although for small α , it still provides the most accurate prediction. One can also verify that the first-order Markov approximation again gives the correct value for the Rabi frequency up to second order in x . A pertinent remark here is that, in this case where $\delta \neq 0$, conditions (24b) and (24c) are rather more complicated to impose than condition (24a). This lends justification to improving the description of the system by going to higher-order Markov approximations using only the simplest condition (24a), instead of correcting the zeroth-order approximation by a more complicated choice of $\tilde{\omega}$.

To further illustrate matters, we examine the predictions for the evolution of the populations in the three states under the different approximations for the case with Rabi frequencies $\Omega_0 = 0.4\Delta$ and $\Omega_1 = 0.3\Delta$, and overall detuning $\delta = (|\Omega_1|^2 - |\Omega_0|^2)/4\Delta$. The results are plotted in figure 2. Standard adiabatic elimination (thin dashed blue lines in the plot), equivalent to the zeroth-order Markov approximation with condition (24a), gives a period for the Rabi oscillation that is about 6% short of the exact value. The zeroth-order Markov solution with condition (24c), on the other hand, gives the exact Rabi frequency, but makes no statement about the population of the intermediate state. In contrast, the first-order Markov approximation, even with the simplest condition (24a), performs very well, and provides a reasonable estimate of the intermediate state population. Note that all the approximations show the effect of the coarse graining: They do not reproduce the high-frequency modulation of the exact solution. This coarse graining is also behind the fact that the population of the ground state, under the first-order Markov approximation, does not start at 1 at time $t = 0$. Instead, it begins with a value that can be thought of as the average over the initial coarse-grained time step, and is consistent with having an initial nonzero population in the intermediate state in compliance with (21).

4.2. Two atoms

For the single three-level atom in Section 4.1, one hardly needs an approximate treatment. Nevertheless, the reduction of the description to the relevant two levels is a helpful simplification and the coarse-grained probabilities thus obtained are often all one needs to know to determine the important parameter range for an experiment. When more levels are involved, a full treatment may no longer be possible or contain too many irrelevant details. As an example of such a more complex situation, we consider two identical three-level atoms with a cascade configuration, where the target state is a Rydberg state — a state with a large principal quantum number and thus large electric dipole moment — and the atoms are so close to each other that a Rydberg blockade [4] happens.

Initially both atoms are in the ground state $|0\rangle$ and we have $|00\rangle = |0\rangle \otimes |0\rangle$ for the initial state of the two-atom system. With the two driving lasers coupling in the same way and with the same strength to both atoms, the two-atom state will be invariant under the permutation of the atoms for all later times. We use a shorthand notation in which, for instance, $|01\rangle$ denotes the state with one atom in the ground state and the other atom in the target state, $|01\rangle = (|0\rangle \otimes |1\rangle + |1\rangle \otimes |0\rangle)/\sqrt{2}$. In total, then, six two-atom states participate in the evolution: $|00\rangle$, $|01\rangle$, $|0e\rangle$, $|1e\rangle$, $|ee\rangle$, and $|11\rangle$.

Referring to this order, the interaction-picture Hamiltonian is

$$H_I = \hbar \begin{pmatrix} \begin{array}{cc|cc} 0 & 0 & \frac{\Omega_0^*}{\sqrt{2}} & 0 \\ 0 & \delta & \frac{\Omega_1^*}{2} & \frac{\Omega_0^*}{2} \end{array} & \begin{array}{cc} 0 & 0 \\ 0 & 0 \end{array} \\ \hline \begin{array}{cc|cc} \frac{\Omega_0}{\sqrt{2}} & \frac{\Omega_1^*}{2} & \Delta + \frac{\delta}{2} & 0 \\ 0 & \frac{\Omega_0}{2} & 0 & \Delta + \frac{3}{2}\delta \end{array} & \begin{array}{cc} \frac{\Omega_0^*}{\sqrt{2}} & 0 \\ \frac{\Omega_1}{\sqrt{2}} & \frac{\Omega_1^*}{\sqrt{2}} \end{array} \\ \hline \begin{array}{cc|cc} 0 & 0 & \frac{\Omega_0}{\sqrt{2}} & \frac{\Omega_1^*}{\sqrt{2}} \\ 0 & 0 & 0 & \frac{\Omega_1}{\sqrt{2}} \end{array} & \begin{array}{cc} 2\Delta + \delta & 0 \\ 0 & \Delta_{\text{RB}} + 2\delta \end{array} \end{pmatrix}, \quad (27)$$

after the usual dipole and rotating-wave approximations have been applied. The energy of the double Rydberg state $|11\rangle$ is shifted by an energy $\hbar\Delta_{\text{RB}}$ due to the strong dipole-dipole interaction between Rydberg levels. When $|\Delta_{\text{RB}}| \gg |\Omega_0|, |\Omega_1|$, the Rydberg blockade mechanism prevents both atoms from being excited to the target state at the same time, and this mechanism can be used to entangle atoms and implement quantum gates [5, 6]. As a typical value of this energy shift, we take Δ_{RB} to be a few times the size of Δ . Since the double Rydberg state $|11\rangle$ is not going to be populated, the goal is a population transfer from the double ground state $|00\rangle$ to the one-Rydberg-atom target state $|01\rangle$, i.e., a transfer between the two states to which the two left columns and the two top rows in (27) refer.

There are two pairs of resonant transitions: $|00\rangle \leftrightarrow |01\rangle$ and $|0e\rangle \leftrightarrow |1e\rangle$. There are thus two different choices of relevant states, either only the two states $|00\rangle$ and $|01\rangle$, or the four states $|00\rangle, |01\rangle, |0e\rangle$ and $|1e\rangle$. Correspondingly, we have a choice between a $6 = 2 + 4$ or a $6 = 4 + 2$ split, as indicated in (27). It is also possible to apply a two-step Markov approximation, i.e. first approximate four states and in a second step approximate the final two states. But this has no additional benefit, it gives the same result as applying the Markov approximation directly to the $6 = 2 + 4$ split.

The plots in figure 3 show the differences between adiabatic elimination (zeroth-order Markov approximation with condition (24a)) and first-order Markov approximation with condition (24a) as well as the impact of the choice of the number of relevant states. As one would expect, the approximation with only two relevant states (bottom plot) includes less information than the one with four relevant states (top plot). Indeed, in the case of two relevant states, the approximations give a smooth curve without the high-frequency components that one sees in the top plot because the states giving rise to these “wiggles” have been eliminated. The plots also confirm that the first-order Markov approximation gives reliable results for both choices of relevant states.

5. Outlook

Although we managed to give answers to all four of the questions asked in Section 2.2, this is not the end of the story. It will be instructive to apply the first-order Markov approximation to other more complex situations where standard adiabatic elimination

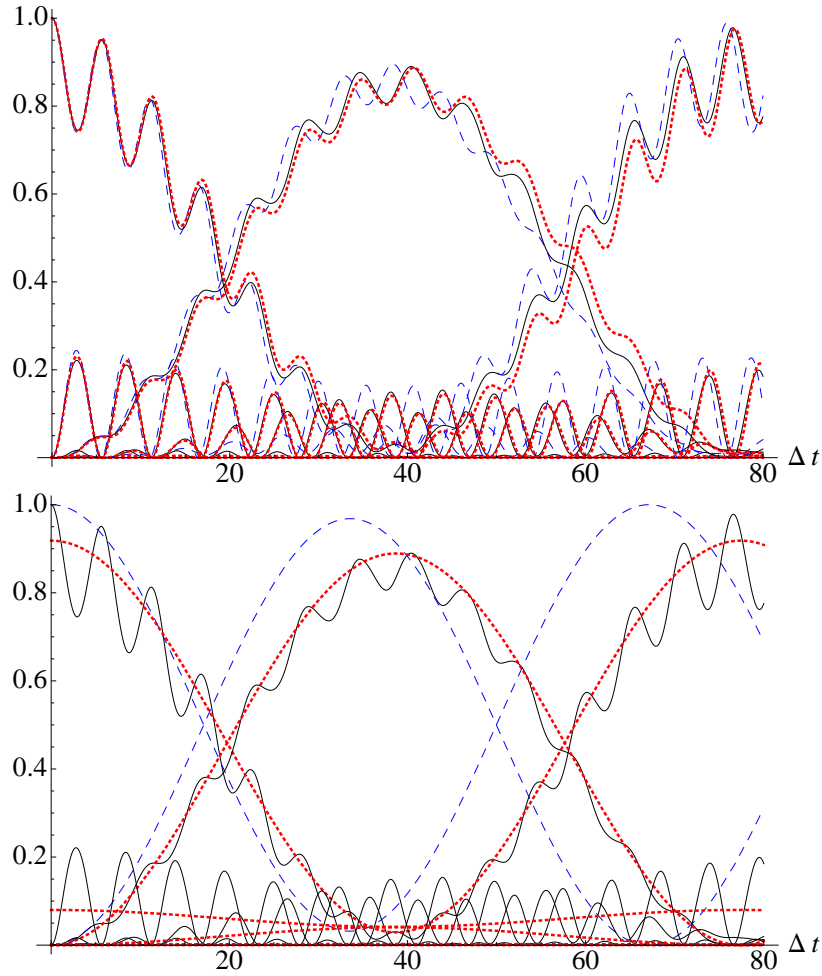


Figure 3. Population probabilities for a two-atom Raman transition in a three-level cascade system with Rydberg blockade, as a function of Δt . The plots are for $\Omega_0 = 0.4\Delta$, $\Omega_1 = 0.3\Delta$, $\delta = 0$ and $\Delta_{\text{RB}} = 5\Delta$, with four relevant and two auxiliary levels in the top plot, and two relevant and four auxiliary levels in the bottom plot. The curves that start near 1 show the probability of the double ground state $|00\rangle$; the curves that start at 0 and increase to about 1 before decreasing again show the probability of the one-atom target state $|01\rangle$; and the curves that start near 0 and never grow to large values show the probabilities of the other states. The black curves show the full solution, the dashed blue curves are for the adiabatic-elimination approximation, and the dotted red curves for the first-order Markov approximation with condition (24a).

is currently the method of choice. As a matter of general interest, the judicious selection of the interaction picture appears to be crucial and deserves further study. One should also extend the method to include spontaneous emission or, more generally, investigate how the method can be modified for open quantum systems. Lastly, we mention that a systematic approximation without the use of adiabatic elimination is possible as well [7].

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